(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) Publication number: **0 375 308 A1**

(12) EUROPEAN PATENT APPLICATION

(21) Application number: 89313184.7

(22) Date of filing: 18.12.89

(51) Int. Cl.5: C22B 21/06, F27D 1/12, F27D 9/00

(30) Priority: 22.12.88 CA 586886

(43) Date of publication of application: 27.06.90 Bulletin 90/26

(84) Designated Contracting States: AT CH DE ES FR GB IT LI SE

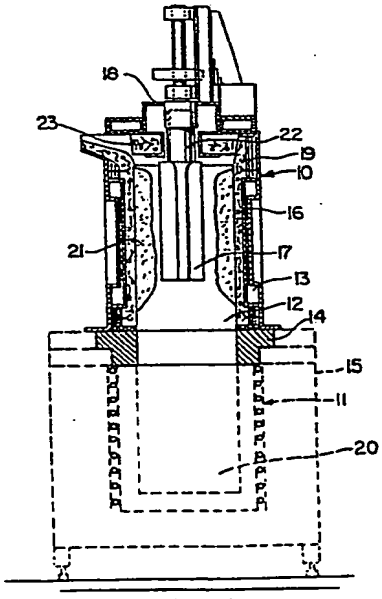
(71) Applicant: ALCAN INTERNATIONAL LIMITED
1188 Sherbrooke Street West
Montreal Quebec H3A 3G2(CA)

(72) Inventor: Celik, Cesur
183 Raddison Pointe-Claire
Quebec G7S 3K9(CA)
Inventor: Dube, Ghyslain
1663 Gay-Lussac
Jonquiere Quebec G7S 3K9(CA)

(74) Representative: Wilkinson, Stephen John et al
c/o Stevens, Hewlett & Perkins 5 Quality Court Chancery Lane
London WC2A 1HZ(GB)

(54) Process and apparatus for producing high purity aluminum.

(57) A process and apparatus are described for producing high purity aluminum by fractional crystallization. This process comprises the steps of melting the aluminum to be purified to obtain a molten aluminum, holding the molten aluminum within a vertical, cylindrical crystallizer vessel at least partially surrounded by a cooling jacket for the passage of cooling fluid therethrough, and stirring the molten aluminum in the vessel while passing cooling fluid through the cooling jacket thereby crystallizing high purity aluminum on the cooled inner vessel wall. An apparatus for carrying out the above process is also described.



EP 0 375 308 A1

Process and Apparatus for Producing High Purity Aluminum

Background of the Invention

This invention relates to a process and apparatus for producing high purity aluminum, particularly by fractional crystallization.

High purity aluminum is widely used to produce automotive and other bright surface products. For instance, a highly desirable aluminum having excellent brightness is one of 99.95% ± 0.02 purity containing less than 0.05 wt% iron and silicon. This high purity aluminum is typically produced by one or a combination of the following techniques: zone refining, three layer refining and fractional crystallization. The zone refining and three layer refining both require high investment, expensive maintenance and very skilled personnel. On the other hand, fractional crystallization tends to be easier to operate, requires minimal investment and is cheaper to run.

The principal objective of fractional crystallization is to form a purified solid phase from an impure liquid phase. When a body of molten aluminum containing eutectic impurities, i.e. Fe, Si, Cu, etc. which undergo a eutectic reaction with aluminum, is subjected to cooling, the crystals which solidify are richer in aluminum content than the liquid body from which they came. It is, therefore, possible by partial solidification of the melt to produce a solid fraction with higher purity than the original melt.

Among specific prior systems for producing high purity aluminum, there may be mentioned that described in Alcoa, U.S. Patent 3,211,547. In the Alcoa method, crystals are formed on the free surface of a molten metal bath and these are allowed to fall to the bottom by gravity to form aggregates of crystals by pressing, thereby separating the crystals from the molten metal. This method suffers from the entrapment of impurities between the crystals collected at the bottom. Further deficiencies are a large diffusion layer between the interface and the liquid and a small temperature gradient.

Reynolds, U.S. Patent 3,163,895 describes a process comprising continuous fractional crystallization of a liquid aluminum feed stream under strong agitation at the solid/liquid interface. The process needs high investment costs and maintenance is difficult and costly.

In Pechiney, U.S. Patent 3,671,229 there is described a system generally similar to that of the Alcoa method, but it is generally not suitable for large scale production and long treatment times are required for small quantities of high purity aluminum.

Mitsubishi, Japanese Patent 1984-28538 describes a system in which molten aluminum is accommodated in a vessel with a horizontal floor. The metal is purified by the extraction of heat through the cooled floor while liquid aluminum is stirred. Cracking problems in the floor and expansion problems with cooling pipes created serious operational difficulties for the Mitsubishi method.

Showa, U.S. Patent 4,469,512 describes a purification method by adding boron to molten aluminum and rotating a cooling body immersed in the boron-containing molten aluminum while introducing a cooling fluid to the interior of the cooling body. This provides a small area of crystallization. Therefore, to increase the surface area, additional vessels identical to each other are set up in series, increasing both the cost and difficulty of the process.

It is the object of the present invention to provide a simpler and less expensive technique for fractional crystallization than has been possible by other methods.

Summary of the Invention

The process of this invention for producing high purity aluminum comprises the steps of melting aluminum containing eutectic impurities to obtain a body of molten aluminum, holding the molten aluminum within a vertical, cylindrical crystallizer vessel, passing cooling fluid through a cooling jacket at least partially surrounding the vessel walls and stirring the molten aluminum within the vessel whereby high purity aluminum crystallizes on the cooled inner surface of the cylindrical vessel.

The molten aluminum is preferably provided in the crystallizer vessel by mounting the crystallizer on top of a melting furnace, e.g. an electromagnetic furnace. The aluminum to be purified is added and melted until the height of the molten aluminum reaches the level of the top of the cooling jacket.

In this manner, eutectic impurities are removed and an annular body of solid, high purity aluminum is formed. The stirring action disperses a layer of molten metal that stays close to the solidification boundary and tends to urge the impurities away from the boundary and into the molten metal. Thus, the impurities do not tend to be trapped in the solids and the purity of the solid product is improved. When the process is

completed, the stirring system is removed, the remaining liquid metal is poured out and the purified solid product is cooled and removed from the reactor.

The aluminum may also contain peritectic impurities, such as vanadium and titanium. If so, an optional prior treatment with boron may be carried out by known methods to remove such impurities.

The methods include the following:

1. Addition of boron as potassium borofluoride;
2. Addition of boron as an aluminium-boron master alloy;
3. Addition of boron by the method disclosed in co-assigned Canadian patent No. 1,215,236.

With the system of the present invention, molten metal temperature, molten metal agitation and the cooling method all affect the quality of the final product. It is found advantageous to use an air/water mist flow for cooling and it has been found that the level of impurities in the purified solid decreases with increased speed of agitation and decreased coolant flow, i.e. lower solidification rate. This coolant flow is obtained by varying the water to air ratio in the coolant mix, either by decreasing the water flow or increasing the air flow.

Best results appear to be obtained with a melt temperature in the order of 660°C ± 0.5°C. Of course, in terms of temperatures, it is a matter of the relationship between the cooling rate and the melt temperature such that the desired freezing rate at the solid/liquid interface is achieved.

During crystal growth, the eutectic impurities are rejected from the solid into the liquid phase to form a boundary layer of concentrated impurities in the vicinity of the liquid-solid interface. As the impurity content in the boundary layer increases, the solid/liquid interface changes from a smooth regular crystal front to a dendritic structure. The thick thermal boundary layer in inefficiently mixed vessels also encourages the growth of dendrites. The dendrites that form at the interface have a tendency to entrap the mother liquid enriched with impurities in their interstices. Therefore their formation is undesirable. To remove this entrapped mother liquid and to reduce the thickness of the thermal boundary layer, strong agitation of the melt is desirable. Thus, agitation of the melt increases the rate of mass transfer because the impurities are more rapidly transferred from the solid/liquid interface to the melt. The higher the rate of agitation, the better is the mass transfer of impurities. Typically, an impeller speed in the order of about 150 to 250 rpm is preferred.

The cooling rate must also be carefully controlled. In order to minimize occlusion of impurities in the crystalline solid, it is desirable to form perfect crystals. However, the more perfect crystal growth occurs at low rates of crystallization. Moreover, as the rate of crystallization is decreased, the rate of buildup of impurities at the solid/liquid interface is reduced. This results in greater separation since decreasing the cooling flow rate decreases the rate of solidification. Of course, there is a lower limit on the level of cooling flow rate and it is must be sufficiently high to produce a rate of heat transfer sufficient to provide the degree of supersaturation necessary for crystallization.

It has been found that a particularly satisfactory result is obtained using as a coolant a mixture of air and water in which a small amount of water is added to an air flow before entering the cooling jacket. This creates a mist flow of very fine droplets of water.

The invention also relates to an apparatus for producing high-purity aluminum comprising (a) a vertical, cylindrical crystallizer vessel at least partially surrounded by a cooling jacket for passage of cooling fluid therethrough and having an open bottom end, (b) a melting furnace connected below the open bottom end of the crystallizer vessel, said furnace being adapted to melt aluminum and provide molten aluminum within the crystallizer vessel and (c) an elongated impeller axially rotatably mounted within the crystallizer vessel, said impeller being adapted to stir the molten aluminum in the crystallizer vessel while passing cooling fluid through the cooling jacket thereby crystallizing high purity aluminum on the cooled inner vessel wall.

Brief Description of the Drawing

The invention will be described below in greater detail with reference to the accompanying drawing.

The drawing is a view in vertical section showing one preferred embodiment of an apparatus for producing high purity aluminum by the process of the invention.

Description of the Preferred Embodiments

The apparatus consists of a cylindrically shaped crystallizer vessel 10 with an open bottom resting on top of an electromagnetic furnace 11. The vessel 10 rests on a support 14 on top of furnace 11 which is

mounted in a cabinet assembly 15. The crystallizer vessel and furnace are both manufactured with silicon carbide refractory walls and the crystallizer vessel 10 has a cylindrical inner chamber 12. A cooling jacket 13 in the form of a hollow wall portion surrounds the vessel 10 and is connected to inlet lines (not shown) for air and water.

Mounted axially within chamber 12 is a graphite impeller 17 with radial vanes. The impeller is driven by a drive shaft 22 extending through removable insulated top 23 and connecting to a motor assembly 18 mounted on top of vessel 10.

Insulation is also provided between the vessel 10 and the electromagnetic furnace 12 and further portions of insulation 19 are provided adjacent portions of the cooling jacket.

In operation, part of the aluminum to be purified is placed within cavity 20 of furnace 11 to be melted and the rest of the aluminum to be purified is added gradually until the height of the molten metal reaches the level of the top of the cooling jacket 13. While stirring the molten aluminum with the impeller 17, the molten metal is cooled by a mist flow of air and water passing through cooling jacket 13. This results in high purity aluminum 21 solidifying on the cooled wall of chamber 12.

The stirring action disperses a layer of molten metal that stays close to the solidification boundary and is rich in impurities away from the boundary and into the molten metal mass. In this manner, the impurities are not pulled into the solids forming on the wall of the vessel and the purity of the final aluminum is improved. When the process has been completed, the stirring system 17, 18 and top 23 are removed and the liquid metal is poured out by tilting the crystallizer using a hydraulic mechanism, leaving an annular form 21 of purified solid which, after cooling, is removed from the vessel 10. The upper half of the crystallizer walls are designed so as to have a few degrees of taper to enable easy removal of the purified solid formed on the walls. Contraction during cooling makes subsequent removal easy. No cutting is necessary.

The reactor described above had the following characteristics:

Crystallizer capacity: 200 kg
Melting furnace capacity: 200 kg
Power capacity during melting: 80 kw
Power capacity to keep the charge at constant temperature: 10 kw
Inside area of the reactor: 6900 cm^2 (1070 in^2)
Height of reactor: 76 cm (30 in.)
Inside diameter: 43 cm (17 in.)
Height of cooling jacket: 51 cm (20 in.)
Width of cooling jacket: 1 cm (3/8 in.)
Maximum flow of cooling air: 7080 liters/minute (250 SCFM)
Maximum flow of cooling water: 1.0 kg/minute

Example 1

The above reactor was used to conduct various experiments on an impure aluminum alloy containing 99.7% by weight aluminum.

(A) In order to study the effects of temperature, tests were conducted at 670°C and 665°C, but very little solidification occurred at these temperatures. To be effective at these temperatures, a greater cooling rate would have been necessary from the cooling jacket. Good solid growth was obtained when the molten metal was kept at 660°C. At this temperature, it was found to be easy to adjust the power of the furnace to keep the molten charge at constant temperature. This consumed about 10 kw of power.

(B) Agitation

For these tests an air flow of 4250 liters/minute was used together with a water flow of 0.40 kg/minute and a melt temperature of 660°C. The rate of agitation was varied and it was found that agitation at speeds below 150 rpm was insufficient for mixing of the melt, while a speed in excess of 250 rpm tended to splash the melt on top of the inside wall of the reactor and cause freezing. The results obtained are shown in Table 1 below:

Table 1

SUMMARY OF EXPERIMENTAL DATA FOR AGITATOR SPEED					
Agitator Speed	Starting Purity	Fe	Si	Ti	V
RPM	%wt Al.	%wt	%wt	%wt	%wt
150	99.76	0.152	0.53	0.0034	0.0081
175	99.76	0.14	0.05	0.004	0.009
225*	99.75	0.15	0.051	0.004	0.008
Agitator Speed	Final Purity	Fe	Si	Ti	V
RPM	%wt Al.	%wt	%wt	%wt	%wt
150	99.92	0.042	0.019	0.012	0.02
175	99.91	0.049	0.02	0.012	0.02
225*	99.92	0.035	0.017	0.014	0.022

*Solidification Rate = 101 kg/hr.
= 1.3 mm/min.

(C) Air Flow

It was found that the degree of purification increases with air flow in the range of 2800 to 5650 liters/min. with constant water flow of 300 g/min. The results are shown in Table 2 below:

TABLE 2

SUMMARY OF EXPERIMENTAL DATA FOR AIR FLOW						
Air Flow	Flow Density	Starting Purity	Fe	Si	Ti	V
liters/min	g water/L	%wt Al.	%wt	%wt	%wt	%wt
2800	0.107	99.77	0.14	0.048	0.003	0.008
4250	0.0705	99.78	0.13	0.048	0.004	0.009
5650+	0.053	99.77	0.14	0.047	0.004	0.009
Air Flow	Final Purity	Fe	Si	Ti	V	
liters/min	%wt Al.	%wt	%wt	%wt	%wt	
2800	99.91	0.050	0.020	0.010	0.019	
4250	99.90	0.050	0.023	0.012	0.021	
5650+	99.92	0.039	0.016	0.011	0.019	

+ Solidification Rate = 86 kg/hr
= 1.2 mm/min.

The effect of decreasing the ratio of water coolant per unit volume of air coolant (col. 2) is to decrease the solidification rate. This tended to improve purification.

(D) Water Flow

The effects of water flow through the cooling jacket were studied using an agitator speed of 150 rpm, an air flow of 4250 liters/minute and a melt temperature of 660°C. The results obtained are shown in Table 3.

TABLE 3

SUMMARY OF EXPERIMENTAL DATA FOR WATER FLOW						
Water Flow	Flow Density	Starting Purity	Fe	Si	Ti	V
kg/min	g water/L	%wt Al.	%wt	%wt	%wt	%wt
0.2	0.047	99.77	0.14	0.049	0.004	0.009
0.3**	0.0705	99.78	0.13	0.048	0.004	0.009
0.4	0.094	99.76	0.152	0.053	0.0034	0.0081
0.6	0.1416	99.76	0.14	0.05	0.005	0.009
Water Flow	Final Purity	Fe	Si	Ti	V	
liters/min	%wt Al.	%wt	%wt	%wt	%wt	
0.2	99.92	0.037	0.017	0.012	0.021	
0.3**	99.90	0.050	0.023	0.012	0.021	
0.4	99.92	0.042	0.019	0.012	0.020	
0.6	99.89	0.060	0.024	0.011	0.018	

** Solidification Rate = 135 kg/hr
= 1.6 mm/min.

The results show that increasing the water coolant flow rate and hence the ratio of water coolant to air coolant (col. 2) increases the rate of heat transfer and tends to decrease the purification of eutectic elements (Fe + Si).

(E) Optimized Conditions

These tests were conducted combining optimized conditions of agitator speed, water flow and air flow. The conditions were an agitator speed of 225 rpm, an air flow rate of 5650 litres/min, a water flow rate of 0.2 kg/min and a melt temperature of 660°C. The results are shown in Table 4 below:

Table 4

SUMMARY OF EXPERIMENTAL DATA FOR OPTIMIZED CONDITIONS					
Test	Starting Purity	Fe	Si	Ti	V
no.	%wt Al.	%wt	%wt	%wt	%wt
1	99.745	0.16	0.06	0.003	0.008
2	99.83	0.08	0.04	0.009	0.01
Test	Final Purity	Fe	Si	Ti	V
no.	%wt Al.	%wt	%wt	%wt	%wt
1	99.93	0.03	0.01	0.01	0.02
2	99.94	0.015	0.01	0.015	0.02

The preferred practice of our invention has by example and otherwise been explained in detail but we do not desire to be limited to such specific description except as expressed in the appended claims.

Claims

1. A process for producing high-purity aluminum comprising the steps of melting the aluminum to be purified to obtain a molten aluminum, holding the molten aluminum within a vertical, cylindrical crystallizer vessel at least partially surrounded by a cooling jacket for the passage of ~~cooling fluid therethrough, and~~ stirring the molten aluminum in the vessel while passing cooling fluid through the cooling jacket thereby crystallizing high purity aluminum on the cooled inner vessel wall..

2. A process according to claim 1 wherein the crystallizer vessel is positioned on top of a melting furnace which provides the molten aluminum for the crystallizer vessel.

3. A process according to claim 2 wherein the stirring is conducted by means of a rotating elongated impeller mounted axially within the cylindrical crystallizer vessel.

4. A process according to claim 3 wherein the impeller is rotated at a rate of about 150 to 250 rpm.

5. A process according to claim 2 wherein peritectic impurities are removed from the molten aluminum by treatment with boron prior to crystallization.

6. A process according to claim 2 wherein the cooling fluid is an air/water mixture.

7. A process according to claim 6 wherein the air/water mixture is a mist flow of very fine droplets of water.

8. A process according to claim 3 wherein upon completion of the process, the impeller is removed from the vessel and an annulus of purified aluminum is removed from the vessel.

9. An apparatus for producing high-purity aluminum comprising (a) a vertical, cylindrical crystallizer vessel at least partially surrounded by a cooling jacket for passage of cooling fluid therethrough and having an open bottom end, (b) a melting furnace connected below the open bottom end of the crystallizer vessel, said furnace being adapted to melt aluminum and provide molten aluminum within the crystallizer vessel and (c) an elongated impeller axially rotatably mounted within the crystallizer vessel, said impeller being adapted to stir the molten aluminum in the crystallizer vessel while passing cooling fluid through the cooling jacket thereby crystallizing high purity aluminum on the cooled inner vessel wall.

10. An apparatus according to claim 9 wherein the impeller has radially extending vanes.

11. An apparatus according to claim 9 wherein the crystallizer vessel has an insulated top with a drive shaft for the impeller extending through said top.

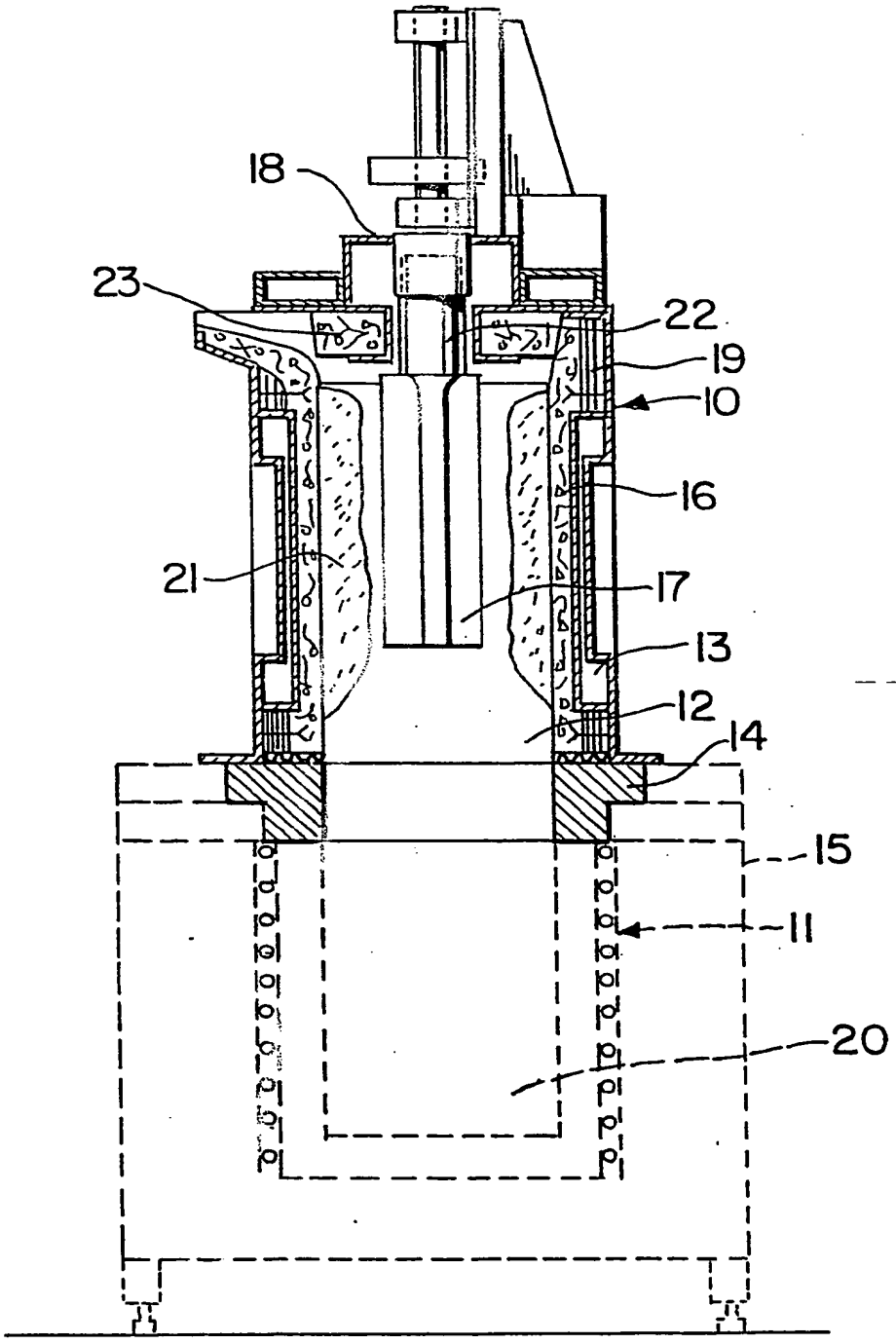
12. An apparatus according to claim 11 wherein a drive motor for the impeller is mounted outside the crystallizer vessel.

13. An apparatus according to claim 9 wherein the cooling jacket comprises a hollow wall portion.

14. An apparatus according to claim 13 wherein the melting furnace is a cylindrical vessel surrounded

by an electromagnetic heater.

15. An apparatus according to claim 11 wherin the top and impeller are removable to permit removal of an annulus of purified aluminum from the vessel.


18
23
22
19
10
16
21
17
13
12
14
15
11
20

European Patent Office

EUROPEAN SEARCH REPORT

Application Number

EP 89 31 3184

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
Y	US-A-4 734 127 (TOSHIAKI IUCHI et al.) * Claims; figure 1; column 1, line 54 - column 4, line 17 *	1,2	C 22 B 21/06 F 27 D 1/12 F 27 D 9/00
A		11,15	
Y	EP-A-0 027 052 (SHOWA ALUMINIUM K.K.) * Figures 1,5; claims 1-3,5-7,9 *	1,2,9	
A		3	
A	PATENT ABSTRACTS OF JAPAN, vol. 6, no. 57 (C-98)[935], 14th April 1982; & JP-A-56 169 736 (SUMITOMO KEIKINZOKU KOGYO K.K.) 26-12-1981 * Whole document *	3,4	
A,D	US-A-3 163 895 (J.L. DEWEY) * Claims; figures 1-2; examples 2-3 *	3,4	
A,D	US-A-4 469 512 (HIDEO SHINGU et al.) * Claims; figure 2 *	6,7	
A	US-A-4 275 569 (R.R. MAYERS et al.) * Figures 5,6; column 2, lines 53-65; claims *	6,7	TECHNICAL FIELDS SEARCHED (Int. Cl.5) C 22 B F 27 D
Y	EP-A-0 064 966 (ISHIZUKA, HIROSHI) * Figure 1; page 3, lines 2-9; claim 1; page 7, lines 21-25 *	9	
A		13,14	

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	22-02-1990	JACOBS J.J.E.G.

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
Y : particularly relevant if combined with another document of the same category
A : technological background
O : non-written disclosure
P : intermediate document

T : theory or principle underlying the invention
E : earlier patent document, but published on, or after the filing date
D : document cited in the application
L : document cited for other reasons

& : member of the same patent family, corresponding document

EPO FORM 1503 03.82 (P0401)